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Influence of the Temperature on Lifetime Reinforcement of a Filled NR

B. Ruellan, J.-B. Le Cam, E. Robin, I. Jeanneau, F. Canévet, and F. Mortier

Abstract Fatigue of crystallizable rubbers has been widely investigated since the pioneer work by Cadwell et al. in 1940 (Cadwell et al., *Ind Eng Chem* 12:19–23, 1940). This study revealed the significant influence of the mean strain on natural rubber lifetime: for non-relaxing loading conditions (*i.e.* $R > 0$), strong lifetime reinforcement was observed and attributed strain-induced crystallization (SIC). Better understanding how SIC reinforces fatigue life is therefore a key point to improve the durability of rubbers. Surprisingly, few studies investigated the effect of temperature on the lifetime, while SIC exhibits a high thermo-sensitivity (Lindley, *Rubber Chem Technol* 47:1253–1264, 1974; Lu *Etude du comportement mécanique et des mécanismes d'endommagement des élastomères en fatigue et en fissuration par fatigue*. PhD Thesis, Conservatoire National des Arts et Métiers, 1991). The present study aims therefore at investigating how temperature affects the fatigue life reinforcement due to SIC under non-relaxing loading conditions. First of all, fatigue tests are carried out at 23 °C. Damage leading to the end-of-life is investigated at both the macroscopic and the microscopic scales for loading ratios from -0.25 to 0.35 . As expected, NR exhibits a reinforcement for positive loading ratios R at 23 °C. Damage, striations due to SIC and number of cycles at crack initiation are mapped using the Haigh diagram. At 90 °C, the fatigue lifetime reinforcement is lower and the signature of SIC on the failure surface disappears. The competition between non-relaxing loading effect and temperature is finally discussed.

Keywords Natural rubber · Fatigue · Strain-induced crystallization · Lifetime reinforcement · Temperature

8.1 Introduction

Literature is very abundant on fatigue of rubbers at ambient temperature.¹ Even though studies differed in the sample geometries tested, in the material formulations and in the loading conditions, general comments can be drawn:

- volumetric samples were generally preferred. Cadwell et al. initially used cylindrical samples [1], then replaced by a diabolo-like geometry introduced by Beatty in 1964 [2]. In the next studies, this type of geometry will be used as the reference one, with a radius of curvature equal to 42 mm [3–16]. Another sample geometries, AE2 and AE5 with lower radius of curvature (2 and 5 mm, respectively), were introduced for investigating multiaxial fatigue of rubber [7–10, 14].
- three end-of-life criteria were used to determine the end-of-life (*i*) the sample failure [1, 15, 17] (*ii*) the occurrence of a crack of a given length at the sample surface. The crack length varies depending on the authors: 1–3 mm [4, 5, 9, 10, 14].

¹In case of strain prescribed test: $R_e = \epsilon_{\min}/\epsilon_{\max}$.

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The choice of a crack length is not physically motivated as discussed in André et al. [7]. Moreover, it is based on an optical detection that is difficult to perform. (iii) a critical value of a macroscopic parameter. This criterion applies for initiation of external as well as internal cracks and is therefore easily applicable to fatigue test. Authors link the end-of-life to a drop in the chosen parameter value: 15% of the maximal effort at the 128th cycle [18], 50% of the stabilized shearing effort [5], 20% of the maximal displacement at the 1000th cycle, for tests under prescribed force [16], etc. Other authors correlated the sample end-of-life with the brutal decrease of the maximal reaction force [3]. As the end-of-life criterion differed from one study to another one, comparing the different studies seems complicated.

- as soon as 1940, Cadwell and co-workers showed the influence of the mean stress on fatigue life of NR [1]. Indeed, a strong reinforcement was observed for loading ratios superior to zero. This was corroborated in Refs. [7, 9]. Such reinforcement was not observed for non-crystallizable rubbers [19, 20], which suggested that SIC is responsible for this lifetime reinforcement.
- for static loadings, even though Lindley suggested that NR would behave as a non-crystallizable rubber for sufficiently high temperature [19], this result cannot be directly transposable to fatigue. Treloar estimates the crystallites melting temperature between 75 and 100 °C [21]. Later, authors showed that SIC had almost completely disappeared (i.e. crystallinity measured close to zero) for experiments carried out at 75 °C, 80 °C or at 100 °C and even for large strains. Under fatigue loadings, several studies investigated the effect of temperature [1–3, 16]. Furthermore, in these studies fatigue tests were carried out at constant loading ratios, except in the work by Bathias and co-workers [22] where authors showed that a lifetime reinforcement was still present at 80 °C but occurred for more important mean stress than at 23 °C. Further investigations including a more exhaustive fatigue tests (i.e. for $R > 0$ where NR exhibits a strong lifetime reinforcement at 23 °C) could provide additional information on the effect of the temperature on crystallizable rubber durability. In his work, Lu showed that NR end-of-life drops by a decade when the temperature increases from 0 to 100 °C [3]. Duan et al. [23] and Neuhaus et al. [16] highlighted the effect of temperature on the NR fatigue properties. On plane samples, Duan et al. showed that the fatigue resistance of NR was only altered for important maximal strains for test carried out at $R_e = 0$ and in the range of temperature from 23 to 90 °C. On diabolo samples, Neuhaus et al. measured a shift of the S-N curves towards the decreasing lifetimes as the temperature increases, independently of the strain applied and for fatigue tests performed at $R_e = -1$, for temperatures from 70 to 100 °C. It is worth noting that NR is highly subjected to self-heating, a difference of 50 °C between the bulk and the surface of diabolo tested has been measured [3, 5]. Thus, self-heating has to be taken into account in order to analyse the effect of the temperature on the rubber fatigue behaviour.

As a conclusion, no study investigates the effect of temperature on the lifetime reinforcement due to SIC by characterizing the damage at the microscopic scale. This is the aim of the present study. The next section presents the experimental setup. Then, results are given and discussed. Concluding remark close the paper.

8.2 Experimental Setup

8.2.1 Material and Sample Geometry

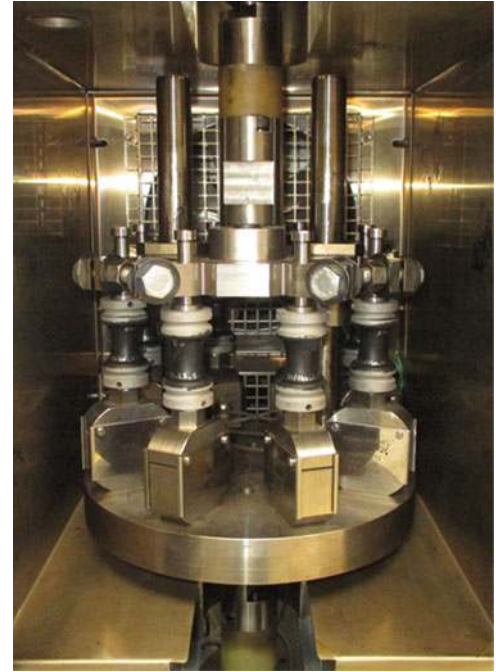
The material considered here is a carbon black filled natural rubber (*cis*-1,4 polyisoprene) vulcanised with sulphur. Samples tested are Diabolo samples.

8.2.2 Loading Conditions

The fatigue tests were performed with a uni-axial MTS Landmark equipped with a homemade experimental apparatus presented in Fig. 8.1. This apparatus enables us to test simultaneously and independently eight Diabolo samples, which compensates the fatigue tests duration and the dispersion in the fatigue life.

The tests were performed under prescribed displacement. The corresponding local deformation at the sample surface in the median zone was calculated by FEA. In order to investigate the effect of temperature on and the fatigue properties, a Servathin heating chamber was used. A pyrometer tracked the temperature of a material point located in the diabolo median surface. For that purpose, a second homemade system (not presented in the figure) has been developed to provide the suitable kinematics to the pyrometer. The frequency was chosen in such a way that the global strain rate $\dot{\epsilon}$ was kept constant, ranging between 1.8 and 2.4 to 2 s^{-1} for one test to another one. In practice, the frequency ranged between 1 and 4 Hz, depending on the strain

Fig. 8.1 Experimental setup. The homemade apparatus with 8 Diabolo samples tested in an oven



amplitude and self-heating to be limited. Five different loading ratios $R_e = \frac{\epsilon_{min}}{\epsilon_{max}}$ were used: -0.25 ; 0 ; 0.125 ; 0.25 and 0.35 . One can recall that loading ratios inferior, equal and superior to zero correspond to tension-compression, repeated tension and tension-tension, respectively.

8.2.3 *End-of-Life Criterion*

Considering a crack initiation approach, a physical criterion based on the maximal reaction force evolution was chosen. Three regimes were obtained during the fatigue test: the softening traduced by a slight decrease of the maximal reaction force, its stabilization and its drop (brutal or not, depending on the loading and environmental conditions). The number of cycles at crack initiation is denoted N_i . It corresponds to the number of cycles at the end of the plateau, when the derivative of N_i is no longer constant. In practice, it corresponds to the occurrence of a macroscopic crack which length is maximum 5 mm at the sample surface.

8.2.4 *Scanning Electron Microscopy*

Second electrons images of diabolo fracture surfaces were stored with a JSM JEOL 7100 F scanning electron microscope (SEM). In addition, the SEM is coupled with an Oxford Instrument X Max Energy Dispersive Spectrometer of X-rays (EDS) and an Aztzec software in order to determine the surface fracture composition, especially in the crack initiation zone. The fracture surfaces to be analysed were previously metallized by vapour deposition of an Au-Pd layer.

8.3 **Results and Discussions**

Results obtained for tests performed at $23\text{ }^\circ\text{C}$ are first described as the reference ones. Then, they are compared to tests carried out at $90\text{ }^\circ\text{C}$ in order to evaluate the influence of the temperature on the material reinforcement. For the both temperatures, lifetime as well as damage mechanisms were considered.

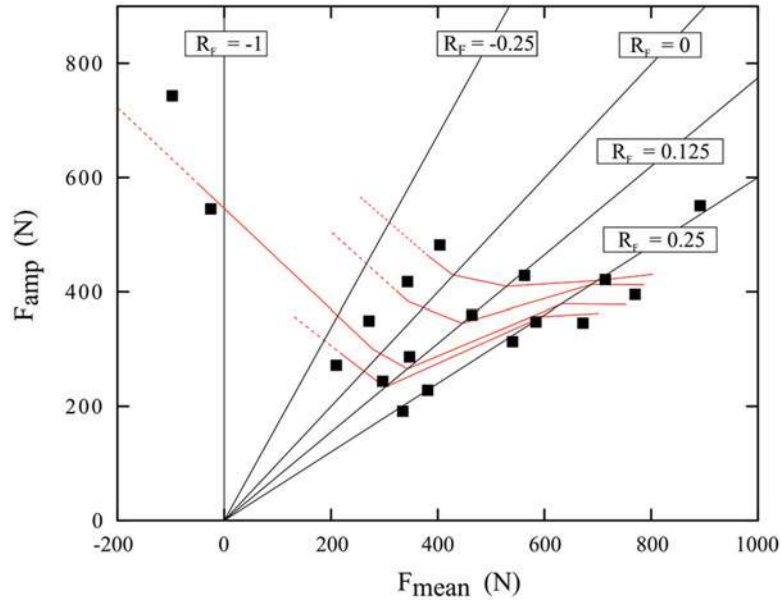


Fig. 8.2 Haigh diagram at 23 °C

Considering tests performed at 23 °C, results are presented in the Haigh diagram plotted in Fig. 8.2. The Haigh diagram provides the iso-lifetime curves in relation to the amplitude and the mean value of the loading. It is classically used to highlight the NR lifetime reinforcement obtained for non-relaxing tension. Here, even though tests were performed under prescribed displacement, the Haigh diagram is plotted in terms of force. Each square in the diagram represents a fatigue test and is linked to a mean lifetime based on eight tests. Considering the loading ratio, three zones can be put into light:

- $-0.25 < R_F < 0$ (tension-compression). The iso-lifetime curves are decreasing monotonously, fatigue damage is driven by F_{\max} . In other words, the mean local strain F_{mean} has no influence on the rubber fatigue life.
- $0 < R_F < 0.125$ (tension-tension). For positive loading ratios, F_{mean} has an influence on the rubber durability: the slope of the iso-lifetime curves increases in absolute value.
- $R_F > 0.125$ (tension-tension). The slope of the iso-lifetime curves becomes positive, exhibiting strong lifetime reinforcement.: at a given F_{amp} , the lifetime increases when F_{mean} is increased. For $R_F > 0.25$, the reinforcement is of a less importance.

Post-mortem analyses have been carried out at both the macroscopic and the microscopic scale on damage mechanisms leading to the sample failure. They enabled us to map fatigue damage in relation to the loading applied. This part of the work is not detailed here.

For fatigue tests carried out at 90 °C, results differ in many point compared to fatigue test at 23 °C. Results are presented in Fig. 8.3 and can be summed up as follows:

- $-0.25 < R_F < 0$ (tension-compression). Similarly to tests at 23 °C, the iso-lifetime curves are decreasing, meaning that F_{\max} drives fatigue damage for this loading condition. Nevertheless, the fatigue lifetime drops by a factor 2 at $R_F = 0$ when the temperature increases from 23 to 90 °C. This result is in good agreement with the literature [16, 23].
- $0 < R_F < 0.25$ (tension-tension). A slight lifetime reinforcement occurs, it is more significant for the shortest lifetime. For similar value of F_{mean} , the reinforcement at 90 °C is not as important as at 23 °C. Nevertheless, reinforcement is still observed and SIC still plays a role in the fatigue behavior at 90 °C, while crystallites are generally assumed to be melt at this temperature level [24].
- $R_F > 0.25$ (tension-tension). Experiments involving such loading conditions are currently in progress and are therefore not presented here.

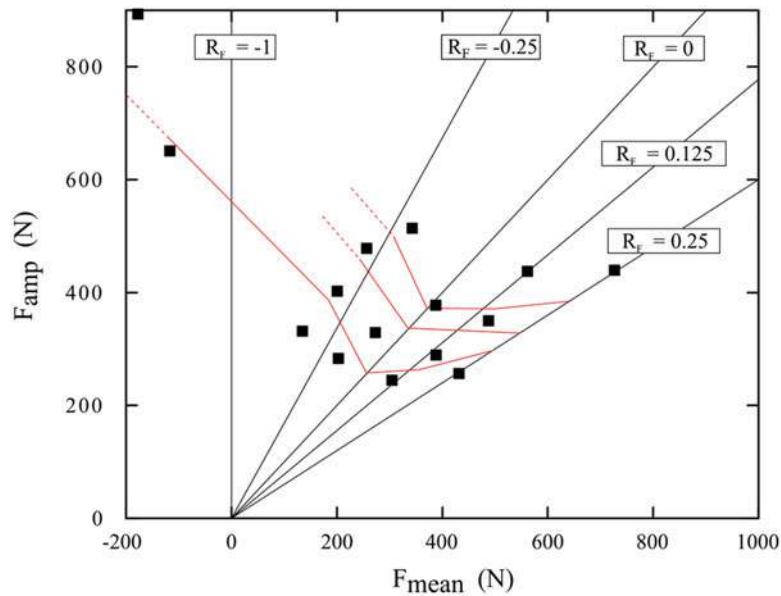


Fig. 8.3 Haigh diagram at 90 °C

8.3.1 Discussion

SIC plays a role of paramount importance in NR fatigue resistance. At 23 °C an important lifetime reinforcement was observed for non-relaxing loading conditions and attributed to SIC. This result was corroborated by the presence of SIC markers on the sample fracture surfaces: wrenchings and fatigue striation. Softer lifetime reinforcement and no SIC signatures were observed at 90 °C, confirming that SIC is strongly altered by the temperature increase. Nevertheless, the reinforcement is still observed. Different conclusions are drawn in comparison to Bathias and co-workers [22] who observed a similar reinforcement at 80 °C and at 23 °C, but for more important mean stress. Even though the crystallinity of filled NR is close to zero for static loading performed at 90 °C, the fatigue loading promote SIC and could be an explanation for the presence of a lifetime reinforcement at 90 °C. It also suggests that only a small value of crystallinity is sufficient to activate the fatigue reinforcement of NR.

8.4 Conclusion

Uni-axial fatigue tests including loading ratios from $-0,25$ to $0,35$ were performed with filled NR at 23 °C and 90 °C. Both lifetime and fracture surface were considered and mapped in Haigh diagrams. Results confirm the high thermosensitivity of NR fatigue properties. Indeed, the material exhibits a strong lifetime reinforcement attributed to SIC at 23 °C. At 90 °C, a softer reinforcement remains for $0 > R_F > 0.25$. This result suggests that during non-relaxing fatigue the crystallinity at 90 °C would not be equal to zero and that only a small value of crystallinity would be sufficient to activate a reinforcement.

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